Production of C60 plasma

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Production of C_{60} plasma

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An ultrafine-particle plasma consisting of electrons, positive K^+ ions, and large negative C_{60}^- ions is produced by introducing "Buckminsterfullerene, C_{60}" particles into a low-temperature ($\simeq$0.2 eV) potassium plasma column confined by a strong axial magnetic field. With an increase in the C_{60}^- fraction, the electron shielding decreases, yielding clear effects on plasma collective phenomena, which are demonstrated for low-frequency electrostatic plasma-wave propagations and instabilities. This plasma might be useful for producing new C_{60}-based materials.

I. INTRODUCTION

Plasmas including dusts or fine particles are of current interest in various fields of physics and engineerings.[1] Under the condition of no direct electron-impact ionization and decomposition, large particles are negatively charged up in plasmas such that there is no net electric current to the particles. Even in case of ultrafine particles, if they are electronegative, i.e., their electron affinity is large, they are negatively charged up in plasmas. In most cases, however, fine particles do not have well-defined mass and size, and thus it is often quite difficult to understand plasmas including such dusts or fine particles.

In this work, "Buckminsterfullerene, C_{60}" particles[2] are used for the purpose of producing a simple plasma including negative ions; C_{60} particles are large cage-like molecules composed of 60 carbon atoms. They are stable, having well-defined mass of mass number $\simeq$720 and size of about 0.7 nm in diameter. In our work, C_{60} particles are introduced into a low-temperature ($\simeq$0.2 eV) plasma column consisting of electrons and potassium K^+ ions, which is produced by contact ionization of K atoms and is confined radially by a strong axial magnetic field in a Q machine.[3] According to careful analysis of ion species, the plasma obtained consists of electrons, K^+ ions, and C_{60}^- ions; C_{60} particles are much smaller than usual fine particles but is much larger than positive ions in our plasma. Preliminary measurements of this ultrafine-particle plasma have been reported at the meetings on plasma physics. Some of them will also appear in Ref. 4.

As is well known, since a simple method was established for producing C_{60} particles in large quantities,[5] a number of works have been carried out to produce new materials composed of C_{60}.[6,7] Our ultrafine-particle plasma might provide a new approach in the field of such a material science.

In Sec. II, experimental apparatus and methods are described. Experimental results are presented, together with discussions, in Sec. III. Section IV contains conclusions.

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II. EXPERIMENTAL APPARATUS AND METHODS

The experiment is carried out in a single-ended Q machine[3,8] with a vacuum chamber, 15.7 cm in diameter and 400 cm long, with pumping systems at both ends. A plasma consisting of electrons and potassium ions K^+ is produced by contact ionization of K atoms at a hot 5.2 cm diameter tungsten plate heated up to 2000 °C under the "electron-rich" condition. The background gas pressure is smaller than 1×10^{-6} Torr. The plasma column, with density $n_p=(1.0\sim5.0)\times10^9$ cm^{-3} and electron temperature $T_e\simeq0.2$ eV$\gtrsim T_+$ (ion temperature), flows along a uniform magnetic field $B\lesssim4$ kG and is terminated by a metal endplate situated at a distance of 210 cm from the hot plate, which is kept at floating potential.

As shown schematically in Fig. 1, a Q-machine plasma passes through a copper cylinder of 7.6 cm in diameter and 20 cm long, situated at a position around the machine center. The cylinder has a 3 cm diameter hole on the side wall, where we set an oven for sublimation of C_{60}. The oven, which is also made of copper, has a small hole to inject C_{60} in the direction toward the Q-machine plasma. The fullerene C_{60}, which is produced by discharge between carbon electrodes in a closed system,[5,9] is kept as a soft film on the inner surface of the oven. The temperature for the C_{60} sublimation is in the range 350~400 °C. The oven temperature T_0, measured at its bottom, is carefully changed to cover this temperature range while the cylinder is kept at a constant temperature around 400 °C. In the region surrounded by the cylinder, the density of C_{60} particles sublimated is in the range $\lesssim10^{13}$ cm^{-3}, depending on T_0, which corresponds to the pressure range $\lesssim10^{-6}$ Torr. These values are estimated from a C_{60} decrease in the oven. In the region outside the cylinder, the density and pressure of C_{60} particles are smaller by one to two orders of magnitude than those inside the cylinder.

Most of plasma parameters are measured by movable Langmuir probes with heating systems to keep their surfaces clean. An "omegatron" analyzer[10] also with heating system, situated behind a small hole of the endplate, is used for analyzing ion species. This analyzer yields signals due to the cyclotron resonances of ion species. There appear peaks in a collector current I_c of the analyzer when a frequency of ap-

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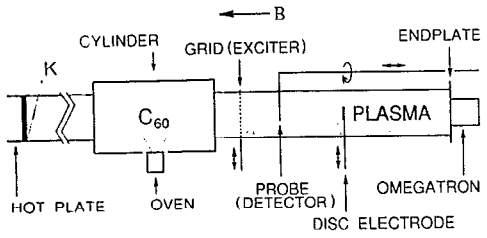

FIG. 1. Experimental apparatus.

plied radio frequency (RF) electric field is equal to the ion cyclotron frequencies. It is also possible to identify the ion species by measuring low-frequency electrostatic plasma-wave propagations and instabilities.[11]

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

The C_{60} control is monitored by measuring a negative-saturation current of the Langmuir probe, I_{s-}, which depends on T_0. A typical variation of I_{s-} is demonstrated in Fig. 2, where the probe is set at the radial center. With an increase in T_0, I_{s-} decreases, implying a decrease in electron density n_e because a change in T_e is not observed. In this experiment, the electron fraction $1-\epsilon$ is found to decrease to the value around 1×10^{-1} at the radial center, where $\epsilon=n_-/n_p$ ($n_p=n_+\simeq n_e+n_-$ if $Z=1$) with positive and negative ion densities, n_+ and n_-, respectively. This value is not as small as in case of SF_6-gas introduction into a Q-machine plasma.[11-13] This is partly because the electron affinity ($\simeq$2.65 eV) is smaller than that of SF_6 ($\simeq$3.39 eV). But, we must be careful about a radial plasma structure modified in the presence of C_{60}^- ions, the Larmor radius ($\simeq$0.46 cm at $B=2$ kG) of which is much larger than that of K^+ ions.

Figure 3 shows typical probe characteristics, i.e., probe current I_p against potential difference V_p applied between the probe and the hot plate which is electrically grounded together with the vacuum chamber (the cylinder and the oven are kept floating), at radial positions (a) $r=0$ cm and (b) $r=1.5$ cm. It can be found in Fig. 3(a) that there also appears a decrease in the positive-saturation current I_{s+} in addition to a decrease in I_{s-} when T_0 is increased. But, I_{s-} decreases

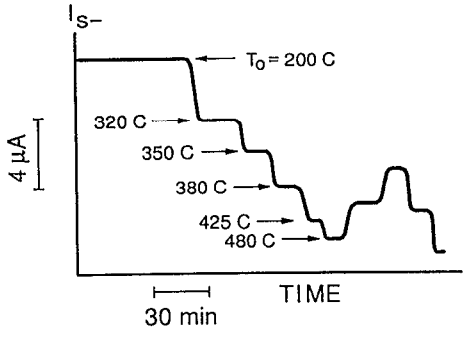

FIG. 2. Control of C_{60} particles: negative probe-saturation current I_{s-} against T_0.

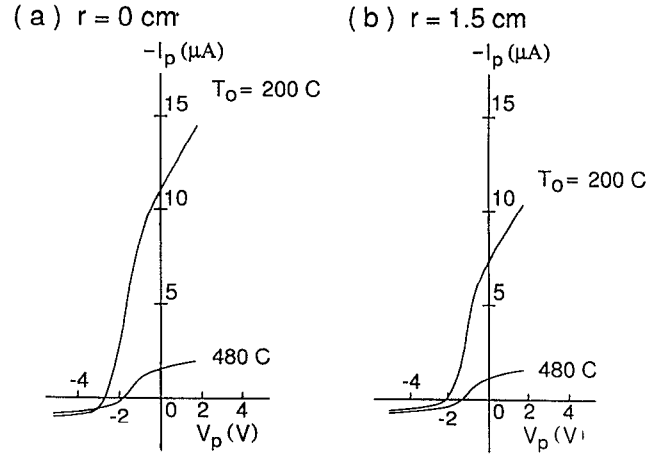

FIG. 3. Typical probe characteristics at (a) $r=0$ cm and (b) $r=1.5$ cm with T_0 as a parameter. $B=2.0$ kG.

more drastically than I_{s+}. This means that the decrease in n_e is accompanied by an increase in n_-. On the other hand, however, at $r=1.5$ cm, with an increase in T_0, I_{s+} increases while I_{s-} decreases, as shown in Fig. 3(b). The increase in I_{s+} implies an increase in $n_p(=n_+)$ at this position. Here it is to be noted that neither plasma cooling nor heating due to the C_{60} introduction can be supposed in our pressure range of C_{60}.

An effect of C_{60} particles on the radial plasma profile is presented in Fig. 4. Since $n_p(=n_+)$ is proportional to I_{s+}, the I_{s+} profile corresponds to a radial variation of the plasma density. On the other hand, I_{s-} includes currents due to electrons and negative ions. The ratio of electron to negative-ion current is given by $(n_e/n_-)(T_e m_-/T_- m_e)^{1/2}$ for electrons of

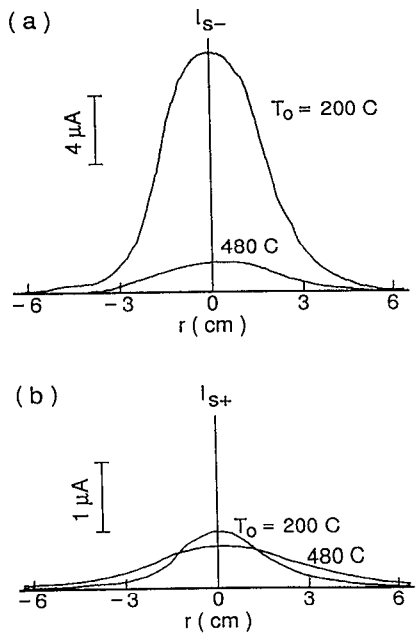

FIG. 4. Radial profiles of (a) I_{s-} and (b) I_{s+} with T_0 as a parameter. I_{s-} and I_{s+} are negative and positive probe-saturation currents, respectively. $B=2.0$ kG.

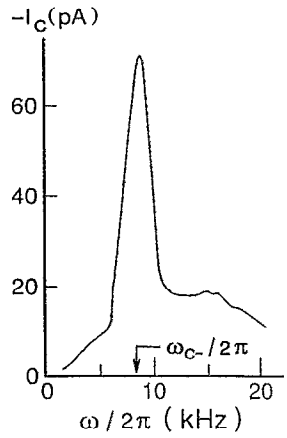

FIG. 5. Collector current I_c of the omegatron as a function of frequency $\omega/2\pi$ of applied RF field at $B=3.85$ kG and $T_0=480$ °C.

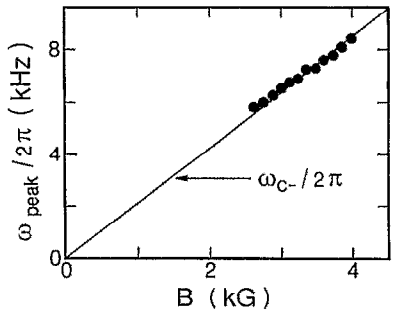

FIG. 6. Frequency at the negative peak in the omegatron spectrum (see Fig. 5), $\omega_{peak}/2\pi$, as a function of B at $T_0=480$ °C. The solid line indicates the predicted cyclotron frequency of C_{60}^-.

mass m_e and temperature T_e and negative ions of mass m_- and temperature T_-. Since this ratio is $1.9\times10^3(n_e/n_-)$ in our case, the negative-ion current is larger than the electron current only when $n_e/n_- \lesssim 5.0\times10^{-4}$ is satisfied, where T_- is assumed to be 0.07 eV because $T_0 \lesssim 500$ °C. In this experiment, the electron current is much larger than the negative-ion current. Thus the I_{s-} profile gives a radial variation of n_e even in the presence of C_{60} particles. As shown in Fig. 4, in the absence of C_{60} particles ($T_0=200$ °C), the I_{s-} profile is almost the same as the I_{s+} profile. By introducing C_{60} particles, the I_{s+} profile becomes broad, showing a plasma spread in the radial direction. This is due to the large Larmor radius of negative C_{60} ions. The I_{s-} profile becomes also broad but is not as broad as the I_{s+} profile, implying that electrons are rather well confined by the magnetic field. Thus, the fraction of negative C_{60} ions increases, i.e., $1-\epsilon$ decreases, resulting in a formation of local plasma structure, in the radial direction. In case of the measurements shown in Fig. 4, we obtain $1-\epsilon \simeq 0.2$ in the central region ($r \lesssim 1.5$ cm) but $1-\epsilon=0.1$ at $r=3.0$ cm, being followed by a further decrease in the plasma edge region.

In order to know how many electrons are attached to the C_{60} particle, ion species are analyzed by using the omegatron analyzer.[10] The collector current of the analyzer, I_c, is plotted as a function of frequency $\omega/2\pi$ of applied RF electric field for cyclotron acceleration. A typical example of the spectra measured is shown in Fig. 5. A clear peak of $-I_c$ is found in addition to the positive peak due to K^+ ions at $\omega_{c+}(=eB/m_+)/2\pi$. A dependence of the frequency giving this negative peak on B is presented in Fig. 6. The results show a good agreement with predicted values of $\omega_{c-}(=eB/m_-)/2\pi$ for C_{60}^- ions. Thus, we can conclude that our negative C_{60} ions are exactly C_{60}^-.

A decrease in n_e gives rise to a decrease in the electron shielding for potential variations. As a result, there appear changes of plasma collective phenomena, which are clearly observed for low-frequency electrostatic plasma-wave propagations and instabilities.[11]

Ion waves are typical low-frequency electrostatic plasma waves. In the presence of negative ions, there are two branches of propagation, i.e., fast and slow modes, corresponding to two ion species.[11,14,15] In our experiment, a grid is used to generate ion waves propagating along a magnetic field toward the downstream region.[8] Small sinusoidal potential variations within 0.1 V peak-to-peak are applied to the grid which is biased a little positively with respect to the floating potential. Typical wave patterns obtained from an interferometer detection system are demonstrated in Fig. 7. It can be found in Fig. 7(b) that there are two modes in the presence of negative C_{60} ions. One of them, the mode with long wavelength (fast mode), propagates even in the absence of C_{60}^- ions, as shown in Fig. 7(a). This mode is due to K^+ ions. The mode with short wavelength (slow mode) propagates only when there are C_{60}^- ions. Figure 8 presents dispersion relations of these two modes, which yield linear relations between $\omega/2\pi$ and k (wave number). This means that the both modes corresponds to ion sound waves with phase velocities $v_p(\simeq\omega/k)$ independent of $\omega/2\pi$.

In a plasma consisting of electrons, positive ions, and negative ions, the phase velocities of the fast and slow modes are given by $(\omega/k)^2 \simeq [T_e/(1-\epsilon)+3T_+]/m_+$ and $(\omega/k)^2 \simeq [\epsilon T_e/(1-\epsilon+T_e/T_+)+3T_-]/m_-$, respectively, where $m_- \gg m_+$ and $T_e \gg T_+ > T_-$ are assumed. In the experiment, $T_e \gtrsim T_+ > T_-$ and there is a flow of K^+ ions, which is due to acceleration by the electron sheath of potential drop $\gtrsim 0.5$ V in front of the hot plate. Thus the above expressions cannot be directly applied to our experiment. But, a qualita-

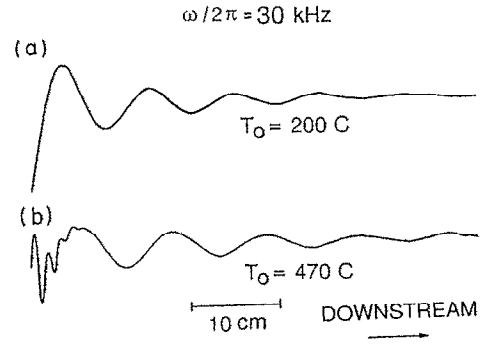

FIG. 7. Wave patterns of ion waves with frequency $\omega/2\pi=30$ kHz at (a) $T_0=200$ °C and (b) $T_0=470$ °C.

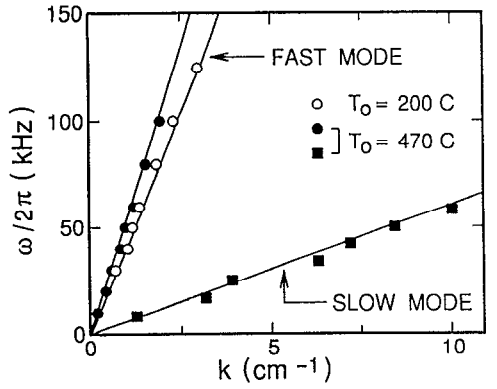

FIG. 8. Dispersion relations of fast and slow ion waves at T_0=200 °C and 470 °C. The best fit lines (straight solid lines) yield the phase velocities v_p=2.5×10^5 cm/s (fast mode, T_0=200 °C), 3.3×10^5 cm/s (fast mode, T_0=470 °C), and 3.8×10^4 cm/s (slow mode, T_0=470 °C).

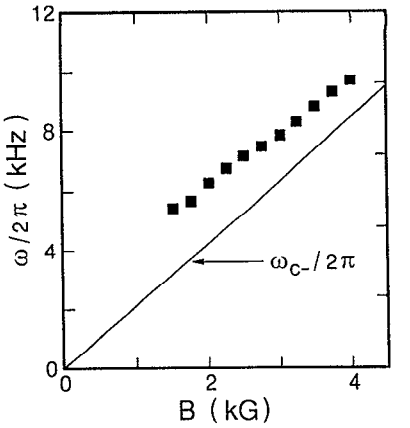

FIG. 10. Frequency $\omega/2\pi$ of ion-cyclotron wave instability due to C_{60}^- ions as a function of magnetic field B at T_0=490 °C.

tive behavior of ion sound waves is well described by these relations. With a decrease in $1-\epsilon$, v_p increases for the both modes, as observed in the experiment. It is also predicted that the fast mode is more sensitive to $1-\epsilon$ than the slow mode. By measuring v_p of the fast mode, we can estimate $1-\epsilon$ even if $1-\epsilon$ is so small that the probe does not give a correct value of $1-\epsilon$.[11]

An example of low-frequency electrostatic instabilities is an ion-cyclotron wave instability.[16] The instability is triggered by applying a positive potential to a small disk electrode situated in front of the endplate. A number of experiments have been performed on this instability,[17] although there are still discussions about the generation mechanism.[18] In the presence of negative ions, there appear two modes of instability[11,13] with frequencies given by $\omega^2 \simeq \omega_{c+}^2 + [T_e/m_+(1-\epsilon)]\kappa^2$ and $\omega^2 \simeq \omega_{c-}^2 + [\epsilon T_e/m_- \times (1-\epsilon+\rho_{+s}^2\kappa^2)]\kappa^2$, where effects of the finite ion Larmor radius are neglected, κ is the wave number perpendicular to the magnetic field, and $\rho_{+s}^2 = m_+ T_e/e^2B^2$. Both of them have the frequencies higher than $\omega_{c+}/2\pi$ and $\omega_{c-}/2\pi$, respectively. Their separations from $\omega_{c+}/2\pi$ and $\omega_{c-}/2\pi$ increase as $1-\epsilon$ is decreased. Figure 9 shows a typical frequency spectrum observed. When C_{60} particles are introduced, there appears a signal due to C_{60}^- ions in addition to the signal due to K^+ ions. The frequency is higher than $\omega_{c-}/2\pi$, as found in Fig. 10, and increases with a decrease in $1-\epsilon$, as predicted theoretically, although the flow of K^+ ions and the finite ion Larmor radius have to be taken into account in order to obtain a quantitative agreement between the measured and predicted values.

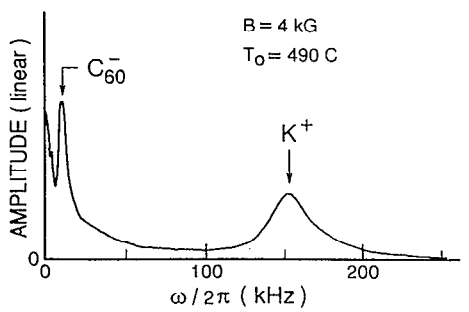

FIG. 9. A typical frequency spectrum of electrostatic ion-cyclotron wave instabilities at B=4.0 kG and T_0=490 °C.

IV. CONCLUSIONS

An ultrafine-particle plasma with C_{60}^- is produced by introducing C_{60} particles into a magnetized low-temperature plasma, where there is no electron-impact ionization and decomposition. The C_{60}^- fraction, n_-/n_+, is controlled in the range $\lesssim$0.9. With an increase in n_-/n_+, the electron shielding effect decreases, yielding clear changes of plasma collective phenomena, as demonstrated for ion-wave propagations and ion-cyclotron wave instabilities.

Because of its simplicity, the plasma produced would be useful for investigating ultrafine-particle plasmas. This plasma might be attractive also in the field of material science. Various knowledges and techniques established in plasma physics and engineerings could be applied to control of C_{60} particles for producing C_{60}-based materials.

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